Mat. Res. Soc. Symp. Proc. Vol. 675 © 2001 Materials Research Society

Endo-fullerenes and Doped Bucky Onions as Seed Materials for Solid State Quantum Bits

[1]Seongjun Park, [2]Deepak Srivastava and [3]Kyeongjae Cho
[1]Department of Chemical Engineering, Stanford University, Stanford, CA 94305-5025
[2]Computational Nanotechnology, NASA Ames Research Center, Moffett Field, CA 94035-1000
[3]Department of Mechanical Engineering, Stanford University, Stanford, CA 94305-4040

ABSTRACT

Two different models for solid-state quantum bits have been investigated. Both are based on the nuclear spin of doped atoms in endo-fullerenes or bucky-onions. ^{1}H or ^{31}P have been tested as suitable dopant atoms because they have half nuclear spins. The thermal stability and electronic properties of the dopant atoms and the encapsulating cages have been examined with *ab-initio* pseudo potential density functional methods, and the results show that both models are suitable for single qubit applications.

INTRODUCTION

Recently a conceptual design of solid-state quantum computer, based on fabricating arrays of ^{31}P atoms in bulk Si, has been proposed. [1] When a ^{31}P donor atom is doped at a substitutional site in bulk Si, four out of five valance electrons of the ^{31}P atom form four tetrahedral covalent bonds with the Si lattice. Kane has argued that the weakly bound fifth electron of ^{31}P donor atom can be used for controlling the nuclear spin state of the donor atom, via hyper-fine interaction, for a solid-state qubit application. The main problem with this proposal, however, is the experimental difficulty involved in fabricating precise arrays of dopant atoms, ^{31}P, in bulk Si layers. Moreover, even if such arrays are fabricated, the individual donor atoms may diffuse away from their intended locations via transient induced diffusion mechanisms.

To overcome the above difficulties, we have investigated encapsulating ½ nuclear spin atoms in endo-fullerenes and bucky-onions for the above described solid-state qubit applications. If the doped endo-fullerene and bucky-onions are stable and have suitable electronic behavior for qubit applications, it may be easier to make the required arrays for solid-state quantum computer applications. The ^{1}H and ^{31}P atoms with ½ nuclear spin have been tried as suitable dopant atoms because both have ½ nuclear spin and one isotope except deuterium ^{2}D. Two different models have been explored for solid-state qubit applications: One model involves encapsulating ^{1}H atom in a fullerene, and the second model involves encapsulating ^{31}P atom in a few nanometer sized diamond nanocrystallite. The fabrication pathway for encapsulating a ^{31}P atom in a diamond nanocrystallite involves first encapsulating a ^{31}P atom in a fullerene at the core of a bucky-onion, and then converting the core to a diamond nanocrystallite by e-beam irradiation and annealing. [2]

All the results in this work are calculated using total energy pseudo-potential density functional theory, [3, 4] where geometry is optimized and the atomic positions are relaxed until the forces become smaller than 0.05 eV/Å.

RESULTS and DISCUSSION

The first model involves using encapsulated ^{1}H atom for a qubit. The ½ nuclear spin state of a bare ^{1}H atom can be a strong candidate to realize a quantum bit, because the atom has one valence electron in 1*s* atomic orbital and hyperfine interaction is fairly strong. For fabrication purpose, however, a bare ^{1}H atom is very reactive and too small to be encapsulate within any molecule or a lattice. Even if there are ways to fabricate arrays with ^{1}H atoms, the atoms are very reactive and form instantaneous chemical bonds with the encapsulating molecules or lattices. The main requirement for encapsulating a ^{1}H atom with in a fullerene is that the encapsulated atoms should stay at the center of the fullerene without forming any bond with the caging fullerene molecule. Fullerenes are the possible candidate for the caging or encapsulating molecules because it is well known that fullerenes have fairly inert internal surface due to pyramidalization. Pyramidalization increases the electron density of the exterior of the surface and decreases the electron density at the interior of the surface due to rehybridization of atomic orbitals in C atom and electron repulsion from σ bonds. [5]

The internal chemical reactivity of fullerenes is examined first. We choose C_{60} and examine the hydrogenation energy. When a ^{1}H atom is introduced at the center of C_{60}, the ^{1}H atom is stable at the center with the formation energy of –0.43 eV. The electron density of the ^{1}H atom at the center shows that the lone 1*s* like valance electron is localized around the ^{1}H atom with a strong hyperfine interaction with the nuclear spin. Though there is no significant or specific binding between the ^{1}H atom and the cage C atoms in C_{60}, the collective sum of small binding interactions between the ^{1}H atom and the cage C atoms make the total binding energy negative. The center site, however, is not found to be the global minimum energy site. The global minimum energy site is found to be due to a chemical bond formation between the ^{1}H atom and a cage C atom of C_{60}. The binding energy for this reaction is found to be –1.23 eV, i.e., much lower than the binding energy of ^{1}H atom at the center. Similar results are found for other fullerenes as well. Generally, the internal surface of a fullerene is less chemically reactive than external surface, but the reactivity is enough for a ^{1}H atom to bind at the cage C atom and not at the center as is desired for the qubit application.

A possible way for reducing the internal chemical reactivity is through the removal of most of the π electron density from the interior of the surface. This can be done through hydrogenation or some other reaction at the exterior of the surface. When the fullerene is fully hydrogenated, C atoms in fullerene have sp^3 atomic hybrid. Because there is no π electron density at the interior of the surface, the reactivity at the interior of the surface is decreased. However, once all the π bonds are removed, the encapsulated ^{1}H atom could escape by diffusion because the C atom ring sizes then increase due to the increased C-C bond lengths. Therefore, a fullerene that is fully hydrogenated on the exterior of the surface, and which has only small number of C atom rings can be a possible encapsulating material. We have investigated dodecahedrane, $C_{20}H_{20}$ for encapsulating ^{1}H atom for qubit application.

The diffusion barrier of ^{1}H atom to escape $C_{20}H_{20}$ as well as the hydrogenation of internal surface of $C_{20}H_{20}$ are examined. The ^{1}H atom is placed at the center of $C_{20}H_{20}$ and moved towards the wall. The changes in the binding energy are calculated at each step. There are 3 high symmetry-binding sites on the internal surface of $C_{20}H_{20}$; top of the C atoms, the center of bonds between the C atoms, and the center of C atom pentagon rings. Binding energy changes along all the three sites are computed as a function of the distance from the center and the results are shown in figure 1. Since there is no π orbital, we confirm that there are no meta-stable

configurations at the interior of the surface. The binding site at the center is the only stable configuration. The diffusion barrier to escape through pentagon ring is also calculated and found to be 1.17 eV. This shows that the system is fairly stable against diffusion at low temperatures. These results are consistent with some other recent experimental [6] and theoretical [7] studies.

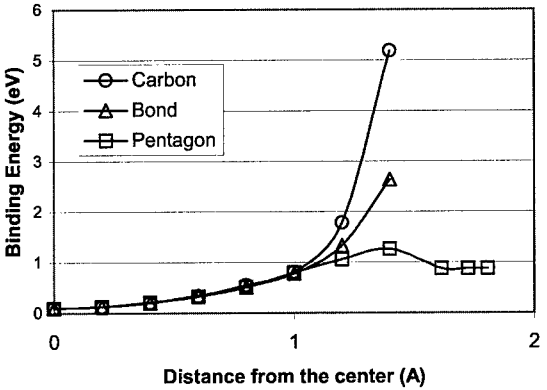

Figure 1. Binding energy of ^{1}H @ $C_{20}H_{20}$:

In order to examine ^{31}P atom in a fullerene as a qubit, the energetically favorable configurations for a ^{31}P atom in a C_{60} fullerene are examined first. For a ^{31}P atom inserted in C_{60}, we identify two energetically stable configurations; in the first case the ^{31}P atom stays at the center of C_{60} , and in the second case the ^{31}P atom is stable on top and at the center of a C-C bond that connects two pentagons on the wall of C_{60}. The binding energies of these configurations are –0.99 eV and –0.81 eV for at the center and on the bond top sites, respectively. The diffusion barrier to escape from the center is also calculated and is found to be 0.33 eV. Therefore, we expect that the ^{31}P atom in C_{60} mostly stays at the center. Recently, there have been experimental observations, which show that ^{31}P atom at C_{60} is stable at the center of the fullerene. [8] This is in agreement with our results. Either of energetically stable configurations, however, does not provide good model for qubit, because valence electrons of ^{31}P atom does not overlap with the nucleus position strongly for a strong hyperfine coupling.

However, a ^{31}P atom encapsulated in a fullerene can be used as a possible first fabrication step in encapsulating the ^{31}P atom in a diamond nanocrystallite. The subsequent steps are: (a) use the encapsulating fullerene to grown bucky onion layers around the fullerenes, and (b) irradiate and anneal the outer layers of bucky onions to convert the core into diamond nanocrystallite doped with a ^{31}P atom. Experimentally, it has been demonstrated that core fullerenes and core layers of buckly onions can be converted to diamond nanocrystallite with controlled e-beam irradiation and annealing. Since a ^{31}P atom at the substitutional site of diamond lattice is similar to ^{31}P atom doped in bulk Si [1], as was suggested by Kane, we have investigated the energetics for possible configurations of ^{31}P doped in a diamond nanocrystallite. The formation energies for possible configurations of ^{31}P atom in normal and compressed (to 20 GPa) diamond lattices, such as, at a substitutional site, a hexagonal interstitial site, and a tetrahedral interstitial site are

calculated, and summarized in table I. The results show that the formation energies for all the configurations are positive, *albeit* a small positive value for the ^{31}P atom at the substitutional site. This shows that there is a small energy cost for inserting a ^{31}P atom at the substitutional site. However, once the ^{31}P atom is trapped (by the above discussed fabrication pathway) at the substitutional site, the diffusion barriers to escape from the nanocrystallite are also very high. Once fabricated, such doped diamond nanocrystallite will be stable, and making arrays of 2-10 nm sized ^{31}P atom doped diamond nanocrystallites in any host material will be much easier than making similar arrays with bare ^{31}P atoms.

Table I. Formation Energy of ^{31}P at different sites of compressed diamond or bulk Si (eV); Formation Energy = E (Si or Diamond: ^{31}P) – E (Si or Diamond) – E (Atomic ^{31}P)

	Diamond			Si
Pressure	0 Pa	20 Pa	50 Pa	0 Pa
Substitution	0.88	1.99	3.85	-5.75
Hexagonal Site	15.95	18.26	21.99	-2.71
Tetrahedral Site	19.13	21.53	25.35	-1.90

CONCLUSIONS

We have discussed two different models for encapsulating nuclear spin ½ atoms for solid-state quantum bit applications. For the first model, we show a way to capture a ^{1}H atom at the center of a fully hydrogenated fullerene, $C_{20}H_{20}$. For the second model, we suggest a way to fabricate diamond nanocrystallite doped with ^{31}P atom at the substitutional site. Making arrays of such larger sized qubits in any host material will be easier than making arrays of bare ^{31}P atoms in bulk Si as was suggested originally. After this work was completed, we became aware about a European collaborative research project, Quantum Information Processing Device using Doped Fullerene, (QIPD-DF) of the similar broad nature. [9] It is very encouraging that this project also aims to apply the fullerene-encapsulated atoms and clusters for novel quantum information device applications. We note that our original research proposal was based on multi-shell fullerenes for solid state quantum computer applications, whereas the QIPD-DF project involves 8 partners with diverse quantum information device processing concepts.

ACKNOWLEDGMENTS

The research performed is supported by NASA Ames Director's Discretionary Fund (DDF) Award to DS (NASA Ames) and KC (Stanford). DS is supported by NASA contract 704-40-32 to CSC. The DFT simulations are performed under the NPACI allocation SUA239 "Nanoscale Materials Simulations."

REFERENCES

1. B. E. Kane, *Nature* **393**, 133 (1998)
2. F. Banhart and P. M. Ajayan, *Nature* **382**, 433 (1996)
3. M. C. Payne, M. P. Teter, D. C. Allan, T. A. Arias and J. D. Joannopoulos, *Rev. Mod. Phys.* **64**, 1045 (1992)
4. S. Ismail-Beigi and T. A. Arias, *Comp. Phys. Comm.* **128**, 1 (2000)
5. A. Hirsch, *Topics in Current Chemistry* **198**, 1 (1998)
6. R. J. Cross, M. Saunders and H. Prinzbach, *Organic Lett.* **1**, 1479 (1999)
7. D. A. Dixon, D. Deerfield, G. D. Graham, *Chem. Phys. Lett.* **78**, 161 (1981)
8. C. Knapp, N. Weiden, K. Kass, K. P. Dinse, B. Pietzak, M. Waiblinger and A. Weidinger, *Molecular Physics* **95**, 999 (1998)
9. Web site of QIPD-DF project is http://planck.thphys.may.ie/QIPDDF/